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The research reported addresses several of the critical issues in the field of ceramic fiber (whisker) / ceramic matrix composites. The overall goal was to understand the complex relationships between processing, interfacial microstructure, and mechanical properties in order to engineer the overall performance of the composites. The composite systems of interest were; (1) SiC whisker / Al_2O_3 matrix composites, (2) beta-Si_3N_4 whisker / Si_3N_4 matrix composites, and (3) SiC continuous fiber / lithium-alumino-silicate matrix composites.

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THE EFFECT OF INTERFACIAL CHARACTERISTICS ON THE MECHANICAL PERFORMANCE OF CERAMIC - CERAMIC COMPOSITES

FINAL REPORT

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A. STATEMENT OF THE PROBLEM STUDIED

The research reported addresses several of the critical issues in the field of ceramic fiber (whisker) / ceramic matrix composites. The overall goal was to understand the complex relationships between processing, interfacial microstructure, and mechanical properties in order to engineer the overall performance of the composites. The major areas that were addressed were:

(1) Composite Processing - Composites were fabricated with controlled fiber (whisker) / matrix interfaces based upon thermodynamic and reaction kinetic considerations.

(2) Interfacial Characterization - The chemical nature of the fiber (whisker) / matrix interfaces was characterized on the molecular level.

(3) Mechanical Property Evaluation - Both micromechanical properties (interfacial shear strength) and macromechanical properties (strength and fracture toughness) were measured.

(4) Correlation of Interfacial Chemistry and Mechanical Properties - Insights were obtained from tasks (1) - (3) describing the mechanical properties in terms of fundamental parameters related to interfacial structure and chemistry.

The above approach provided the knowledge for the tailoring of interfacial reactions, thus allowing the processing of interfaces with optimum activation of strengthening and toughening mechanisms. Fundamental parameters which maximize crack arrest and deflection and fiber (whisker) pullout and bridging were determined.

B. SUMMARY OF THE MOST IMPORTANT RESULTS

(1) SiC Whisker / Al_2O_3 Matrix Composites

For the SiC whisker / Al_2O_3 matrix composites, two types of commercially available whiskers were evaluated; Silar SC-9 whiskers (Arco Metals Co.) and Tateho SCW-1S whiskers (Tateho Chemical Ind.). The processing procedures to produce dense composites with uniform whisker distribution were established. Composites produced with Silar SC-9 whiskers exhibited enhanced fracture toughness (9.0 $MPam^{1/2}$) compared to unreinforced Al_2O_3 (4.0 $MPam^{1/2}$). These same composites also exhibited rising R-curve behavior, indicative of crack wake toughening mechanisms. Composites produced with Tateho SCW-1S whiskers exhibited fracture toughness levels similar to the unreinforced Al_2O_3 (4.5 $MPam^{1/2}$). The strengths for composites produced with both types of whiskers were in excess of 600 MPa. Examination by SEM of crack / microstructure interactions revealed that crack deflection and whisker pullout and bridging toughening mechanisms were operative in the composites produced with Silar SC-9 whiskers. Similar toughening mechanisms were absent in the composites produced with the Tateho SCW-1S whiskers. Whisker surface analysis by XPS and whisker / matrix interface examination by TEM revealed that a thin layer of SiO_xC_y and free C was present on the surfaces of the Silar SC-9 whiskers, while the surfaces of the Tateho SCW-1S whiskers were relatively clean. For the composites produced with the Silar SC-9 whiskers, it is believed that this difference in whisker surface chemistry prevented a strong chemical bond at the interface and was responsible for the activation of the toughening mechanisms sited.

In an effort to further enhance the fracture toughness of the composites produced with the Tateho SCW-1S whiskers, whisker surface modification experiments were performed. Whiskers were treated at elevated temperatures in air, H_2/N_2, and H_2/Ar atmospheres. The air treatment increased the SiO_2 species on the surface of the whiskers. Both the H_2/N_2 and H_2/Ar treatments increased the free C species on the surface. No increase in fracture toughness was found for the composites produced with the air treated whiskers. The composites produced with the H_2/N_2 and H_2/Ar treated whiskers exhibited increases in fracture toughness of 5.3 and 6.3 $MPam^{1/2}$, respectively. Although the presence of free C species enhanced the toughness, the presence of the SiO_xC_y phase appeared to be a more important factor.

This investigation clearly demonstrated that the surface chemistry of the SiC whiskers had a major impact on the mechanical performance of the composites. Fracture toughness ranged from 4.0 to 9.5 $MPam^{1/2}$, depending on the surface species present. The nature of the surface species appeared to affect the whisker / matrix interfacial bonding and thus the extent of the crack / microstructure interactions. The presence of SiO_xC_y or free C surface species was associated with the higher fracture toughnesses. Crack / microstructure interactions, including whisker pullout and crack deflection along whisker / matrix interfaces, were observed in these composites. The presence of SiO_2 or the absence of surface species was associated with the composites that exhibited the lower fracture toughnesses. In these cases, little or no crack / microstructure interactions were observed.

(2) Beta-Si_3N_4 Whisker / Si_3N_4 Matrix Composites

For the beta-Si_3N_4 whisker / Si_3N_4 matrix composites, a new whisker not yet commercially available was evaluated; UBE-SNW whiskers (UBE Chemical Co.). The beta-Si_3N_4 whiskers were found to be of extreme crystallographic perfection and dimensionally straight, indicating that they are excellent candidates for toughening ceramic matrices. Thermodynamic simulations were performed for processing the beta-Si_3N_4 whisker / Si_3N_4 matrix composites and for modifying the beta-Si_3N_4 whisker surface with BN coatings. For composites fabricated with the umodified beta-Si_3N_4 whiskers, strengths up to 660 MPa and fracture toughnesses up to 8.6 $MPam^{1/2}$ were measured. BN coatings were successfully applied by a vapor deposition process. Composites with the modified whiskers were also fabricated. The application of BN to whisker surfaces was found to be an effective means for activating toughening mechanisms. The composites fabricated with the BN modified whiskers exhibited excellent mechanical properties; strengths approximately 650 MPa and fracture toughnesses approximatley 10 $MPam^{1/2}$.

(3) SiC Continuous Fiber / Lithium-Alumino-Silicate Matrix Composites

For the SiC continuous fiber / lithium-alumino-silicate matrix composites, a commercially available fiber was evaluated; NICALON fibers (Nippon Carbon Co.). To predict the formation of the carbon interphase, thermodynamic calculations were performed. These results indicated that under certain conditions carbon species are stable, along with CO and SiO gas species. Variables in the thermodynanic calculations included temperature, pressure, and composition. Processing procedures were established to control the extent of carbon interphase development by varying the hot pressing parameters. It was found that the extent of the carbon interphase development could be controlled by varying the following hot pressing parameters; the temperature (1000 to 1200°C) and time (0.25 to 4 hours). The measured carbon interphase thickness increased with both temperature and time, ranging from approximately 1400 to 5400 Å.

Fitting the carbon interphase thickness versus time data at each temperature to both a diffusion controlled reaction rate model and a diffusion controlled cylindrical model revealed slopes of approximately 1/2. This indicated that the formation of the interfacial carbon layer involved a diffusion controlled process. The measured activation energy for diffusion corresponded to 25.3 kcal/mole. The magnitude of this activation energy corresponds to the diffusion of gas through a solid. In this case, the diffusion of O_2 or CO could be the rate controlling step. The presence of the predicted gas species was confirmed with thermal analysis techniques; simultaneous thermal analysis with mass spectrometry. From these results, a mechanism for the formation of the carbon interphase was proposed.

C. LIST OF ALL PUBLICATIONS AND TECHNICAL

(1) J. Homeny and W. L. Vaughn, "Whisker-Reinforced Ceramic Matrix Composites", Materials Research Society Bulletin, Vol. 12, No. 7, pp. 66-71, 1987. (Reprint)

(2) J. Homeny, W. L. Vaughn, and M. K. Ferber, "Silicon Carbide Whisker / Alumina Matrix Composites: Effect of Whisker Surface Treatment on Fracture Toughness," Journal of the American Ceramic Society, Vol. 73, No. 2, pp. 394-402, 1990. (Reprint)

(3) W. L. Vaughn, "Processing and Characterization of Silicon Carbide Whisker/Aluminum Oxide Matrix Composites", Ph.D. Thesis, University of Illinois, December 1987. (Thesis)

(4) J. Homeny, W. L. Vaughn and M. K. Ferber, "R-Curve Behavior in a SiC Whisker/Al_2O_3 Matrix Composite", submitted to Journal of the American Ceramic Society . (Manuscript)

(5) J. Homeny, L. J. Neergaard, K. R. Karasek, J. T. Donner, and S. A. Bradley, "Characterization of β-Silicon Nitride Whiskers," Journal of the American Ceramic Society, Vol. 73, No. 1, pp. 102-105, 1990. (Reprint)

(6) J. R. Vanvalzah, "Processing, Interfacial Characterization, and Mechanical Property Evaluation of SiC Fiber / Li_2O-Al_2O_3-SiO_2 Glass Matrix Composites," September 1988. (Thesis)

(7) L. J. Neergaard and J. Homeny, "Mechanical Properties of Beta-Silicon Nitride Whisker / Silicon Nitride Matrix Composites," Ceramic Engineering and Science Proceedings, Vol. 10, No. 9-10, pp. 1049-1062, 1989. (Reprint)

(8) J. Homeny, J. R. VanValzah, and M. A. Kelly, "Interfacial Characterization of SiC Fiber / Li_2O-Al_2O_3-SiO_2 Glass Matrix Composites," submitted for publication in the Journal of the American Ceramic Society. (Manuscript)

(9) J. Homeny and L. J. Neergaard, "Mechanical Properties of β-Si_3N_4 Whisker / Si_3N_4 Matrix Composites," submitted for publication in the Journal of the American Ceramic Society. (Manuscript)

(10) J. Homeny and L. J. Neergaard, "Processing, Characterization, and Properties of Beta-Silicon Nitride Whisker / Silicon Nitride Matrix Composites," submitted for publication in the Proceedings of the 5th Technical Conference of the American Society of Composites. (Manuscript)

D. <u>LIST OF ALL PARTICIPATING SCIENTIFIC PERSONNEL SHOWING ANY ADVANCED DEGREES EARNED BY THEM WHILE EMPLOYED ON THE PROJECT</u>

(1) <u>Graduate Students</u>

(1) Janet R. Vanvalzah-M.S. (September 1988)
(2) Mark A. Kelly-Ph.D. (To be completed in May 1991)
(3) Wallace L. Vaughn (partial support)-Ph.D. (November 1987)
(4) Lynn J. Neergaard (partial support)-Ph.D. (To be completed in May 1990)

(2) <u>Undergraduate Students</u>

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(2) Gregory Zeigler
(3) Kyle Jeray
(4) Mark Teepe
(5) Kevin Wesling